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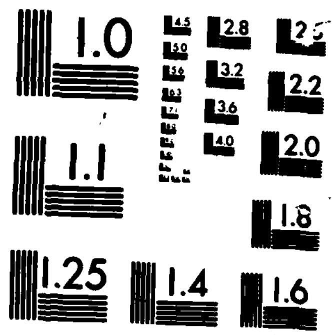

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REPORT NO. SD-TR-86-10

AD-A166 358

Total Inversion on the Bi(6^2D-6^4S) Transition at 8758 Å by Near-Resonant Collisional Transfer with NF($a^1\Delta$)

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28 February 1986

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This report has been reviewed by the Public Affairs Office (PAS) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nationals.

This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

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Total population inversion between the two lowest electronic states of atomic bismuth, (6^2D) and (6^4S), has been demonstrated in a subsonic flow system. The atomic inversion is generated via collisions of the ground state $Bi(6^4S)$ with electronically excited $NF(a^1\Delta)$ in the extremely fast resonant transfer process.

$$NF(a^1\Delta) + Bi(6^4S) \rightarrow NF(X^3\Sigma^-) + Bi(6^2D) + 16\ cm^{-1}$$

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20. ABSTRACT (Continued)

This result proves unequivocally that the $NF(a^1\Delta)$, produced by the reaction

$$H + NF_2 \xrightarrow{\text{yields}} HF(v) + NF(a^1\Delta)$$

is also totally inverted. Keywords;

see D[illegible]

PREFACE

The authors thank H. Bixler and R. Cook for their assistance in facility construction; D. Durran for construction of the dc discharge source; and to D. Sutton and D. Spencer for their helpful discussions.

CONTENTS

FIGURES

I. INTRODUCTION

In the search for a short-wavelength laser, the transfer and scaling processes in the NF-Bi system are being investigated.

Excited nitrogen fluoride, $NF(a^1\Delta)$, is a radical with a relatively long radiative lifetime and is somewhat immune to chemical reaction or collisional deactivation. Further, nitrogen fluoride can be efficiently produced by chemical reaction as proposed by Herbelin and Cohen[1,2] and corroborated by Cheah, Clyne, and Whitefield[3] as well as Malins and Setser.[4] The resonant collisional transfer of $NF(a^1\Delta)$ with $Bi(6^4S)$ is fast and efficient as shown by Capelle, Sutton, and Steinfeld.[5] These properties suggest that $NF(a^1\Delta)$ could be employed as a pumping medium in a transfer laser system.

II. EXPERIMENTAL

Our experiments were conducted on the 10-cm flow-tube facility shown in Fig. 1. The fluorine atoms were produced by the dissociation of NF_3 in a dc discharge with an argon diluent. Thermal dissociation of N_2F_4 in an argon diluent at 600°K generated NF_2 according to process 2. Combining fluorine atoms with NF_2 in the presence of hydrogen gives rise to $NF(a^1\Delta)$ according to reactions (3) and (4).

$$NF_3 + dc \rightarrow F + N_2 + \text{side products} \quad (1)$$

$$N_2F_4 + Q \rightarrow 2NF_2 \quad (2)$$

$$F + H_2 \rightarrow HF(v) + H \quad (3)$$

$$H + NF_2 \rightarrow NF(a^1\Delta) + HF(v) \quad (4)$$

Bismuth atoms were introduced into the nitrogen fluoride flow through vaporization in a heater, as shown in Fig. 2. A boat, containing bismuth metal, was fabricated from 1/2-in. stainless steel tubing, pinched at one end, and cut along its length forming a 1 x 6 cm opening. A heating element, capable of operating up to 200 W and 600°C, vaporized the bismuth metal. This assembly is contained within a 1-in. diameter stainless steel cylinder. The quantity of bismuth atoms emitted was regulated by temperature control and flow rate of the helium carrier. The mixture consisted of monomers and dimers in a 2:5 ratio at our nominal run condition of 400°C.[6] The flow tube velocity was maintained at 30 m/sec at a pressure of approximately 2.5 Torr.

The action of $NF(a^1\Delta)$ upon $Bi(6^4S)$ to produce $Bi(6^2D)$ was observed through the two ports located at 15 and 140 cm downstream of the bismuth heater. The $Bi(6^4S)$ concentration was monitored by observing the absorption of the 3068 Å emission[7] from a hollow cathode lamp source with a modified 1/4-m monochromater. Concurrently, the emission from $Bi(6^2D)$ at 8758 Å was

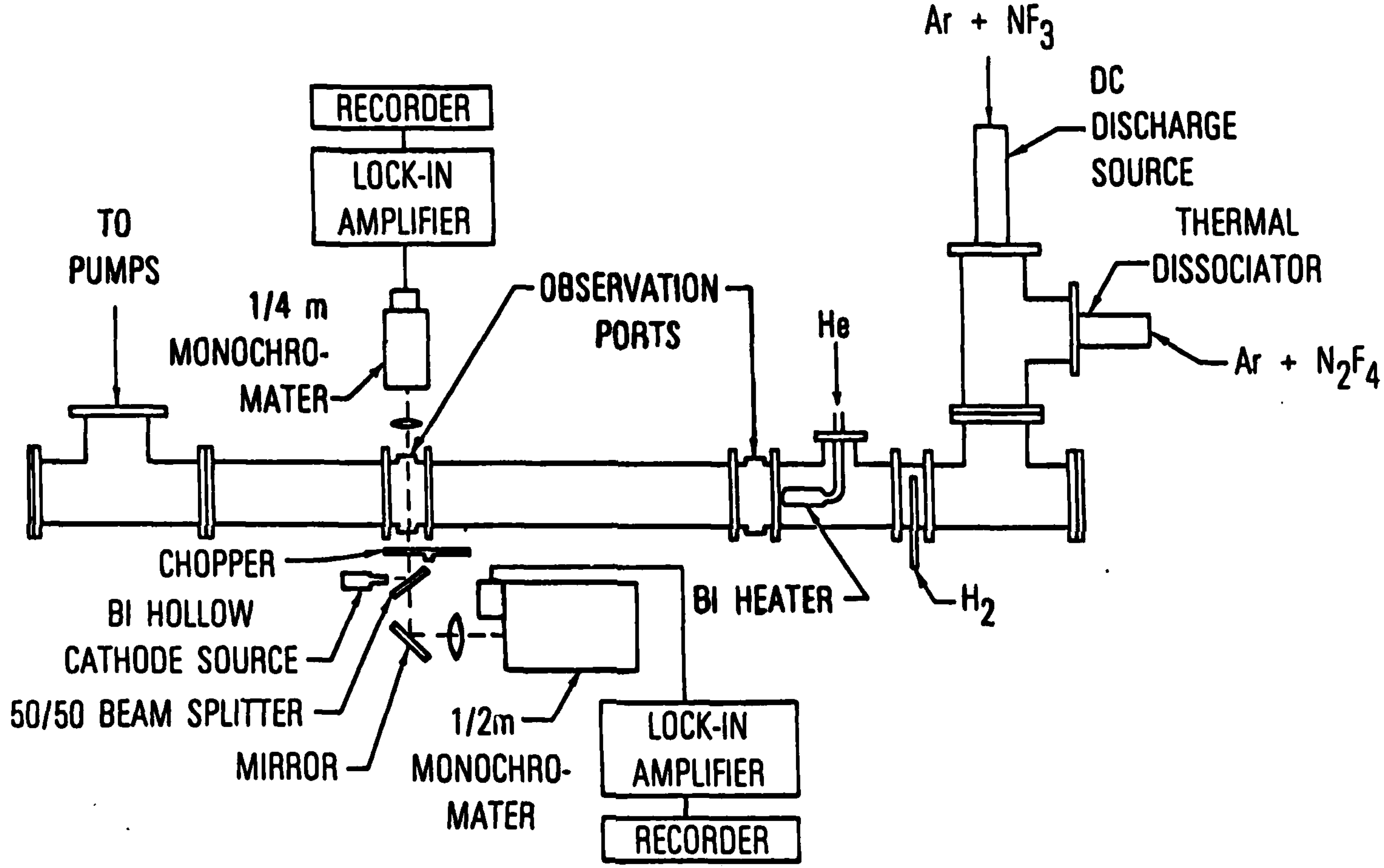

Fig. 1. Schematic of the 10-cm Flow Tube Facility

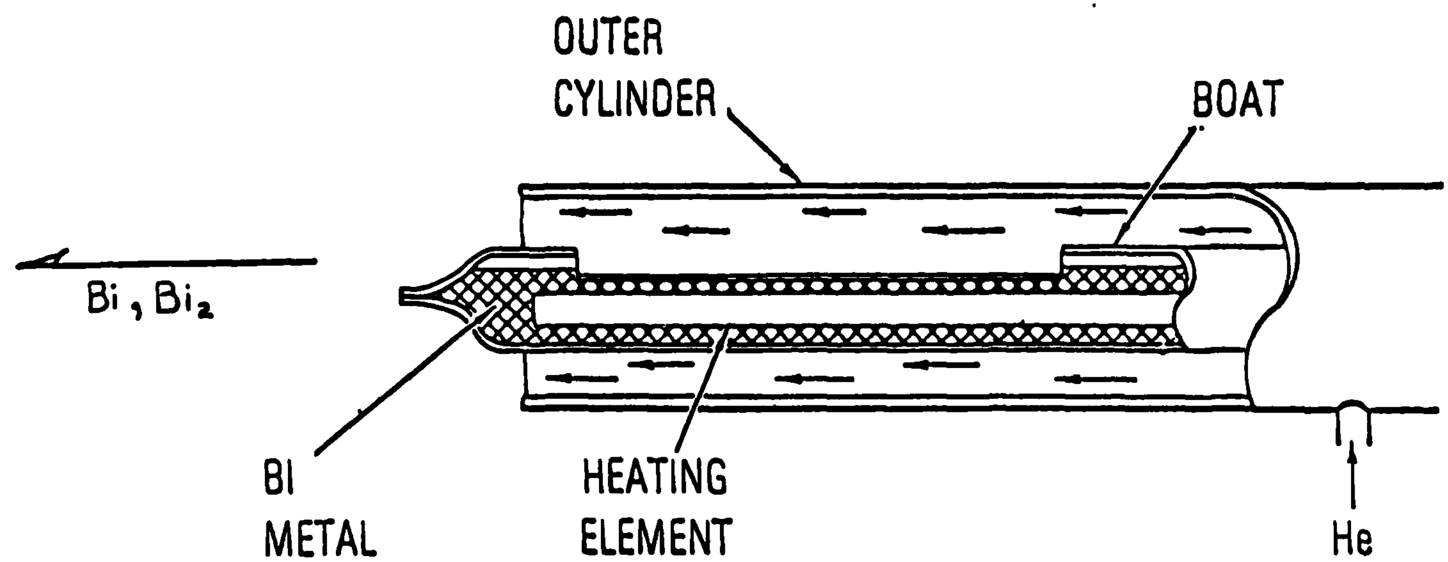

Fig. 2. Cutaway Drawing of the Bismuth Heater

monitored using a 1/2-m monochromater. The concentrations of $NF(a^1\Delta)$ and $Bi(6^2D)$ were computed using the radiative lifetimes of 7 sec for $NF(a^1\Delta)$ and 0.040 sec[8] for bismuth. The $Bi(6^4S)$ concentrations were computed following the method of Trainor[9] using a stimulated absorption cross-section $\sigma = 7.7 \times 10^{-12}$ cm^2, which is computed from the radiative lifetime quoted in Ref. 7.

III. RESULTS AND DISCUSSION

The variation of $NF(a^1\Delta)$ as a function of the N_2F_4 flow rate up to a concentration of about 1.0×10^{13} molecules/cm^3 at the upstream port is shown in Fig. 3. The concentration at the downstream port is significantly lower.

The injection of bismuth atoms into the flow produces $Bi(6^2D)$ by the collisional process (5),

$$NF(a^1\Delta) + Bi(6^4S) \rightarrow Bi(6^2D) + NF(X^3\Sigma^-) + 16\ cm^{-1} \quad (5)$$

which produces the emission spectrum shown in Fig. 4. The $NF(a^1\Delta)$ emission at 8742 Å and the $Bi(6^2D)$ emission at 8758 Å are readily resolved by our instrumentation.

The observed effect of $NF(a^1\Delta)$ upon the $Bi(6^4S)$ at the upstream port are illustrated in Figs. 5a, 5b, and 5c. The graphs reflect typical data for three different initial $Bi(6^4S)$ concentrations in ascending order. In each case, the ground state concentration diminishes as the $Bi(6^2D)$ concentration increases in relation to the N_2F_4 flow rate. For cases in Figs. 5a and 5b, all the available bismuth atoms appear to be pumped, as reflected by the leveling off at the high flow rate end of the excited state curve. Inversion between the $Bi(6^2D)$ and $Bi(6^4s)$ states of greater than 90% appear achievable. Such is not the case for the higher $Bi(6^4S)$ concentration, see Fig. 5c.

The total bismuth atom concentration observed can be obtained by summing the excited and ground state concentrations at each N_2F_4 flow condition. Typically, there is an immediate drop in the total atom concentration, followed by a rise to some maximum valve as seen in Fig. 5a. A loss of bismuth atoms from the excited $Bi(6^2D)$ state could explain the initial drop in total atom concentration. The subsequent rise in total atom concentration is believed to be the result of dissociation of bismuth dimers.

In Fig. 6 are shown the observed maximum $Bi(6^2D)$ concentrations for the initial $Bi(6^4S)$ concentrations of Figs. 5a, 5b, and 5c. The excited state concentration rises quickly to about 6×10^{10} atoms/cm^3, but an additional

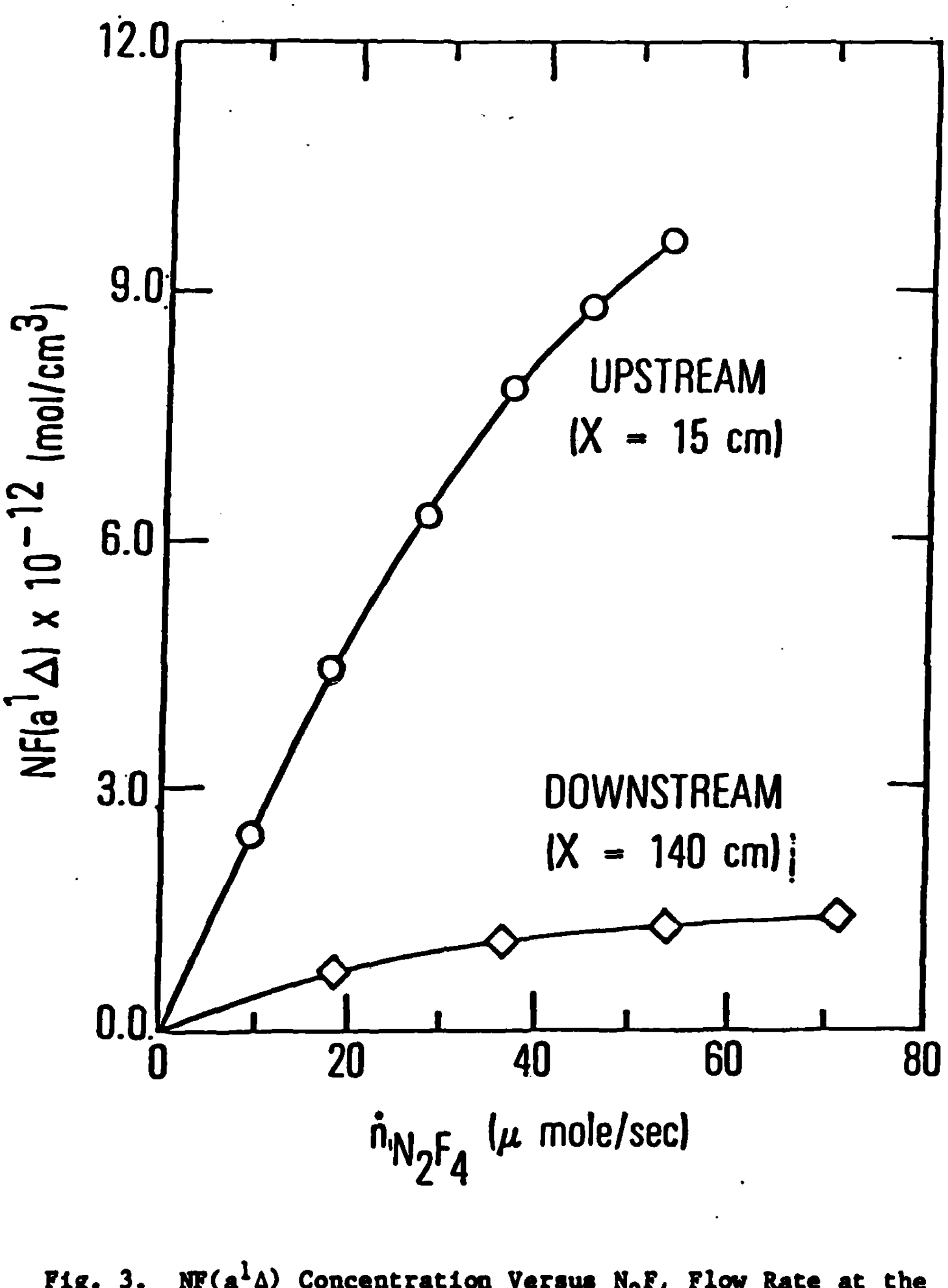

Fig. 3. $NF(a^1\Delta)$ Concentration Versus N_2F_4 Flow Rate at the Two Observation Ports

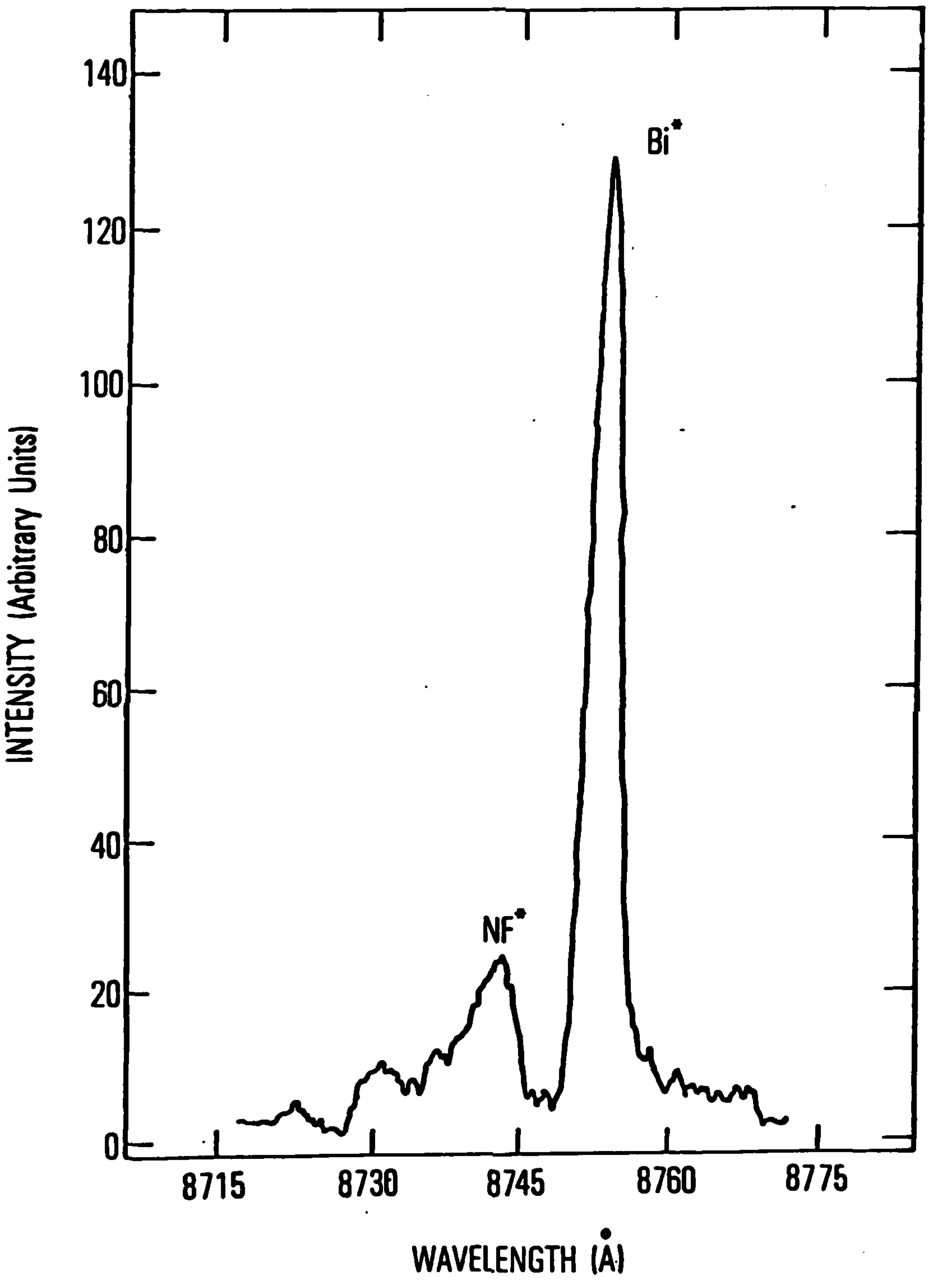

Fig. 4. Emission Spectrum of $NF(a^1\Delta)$ and $Bi(6^2D)$

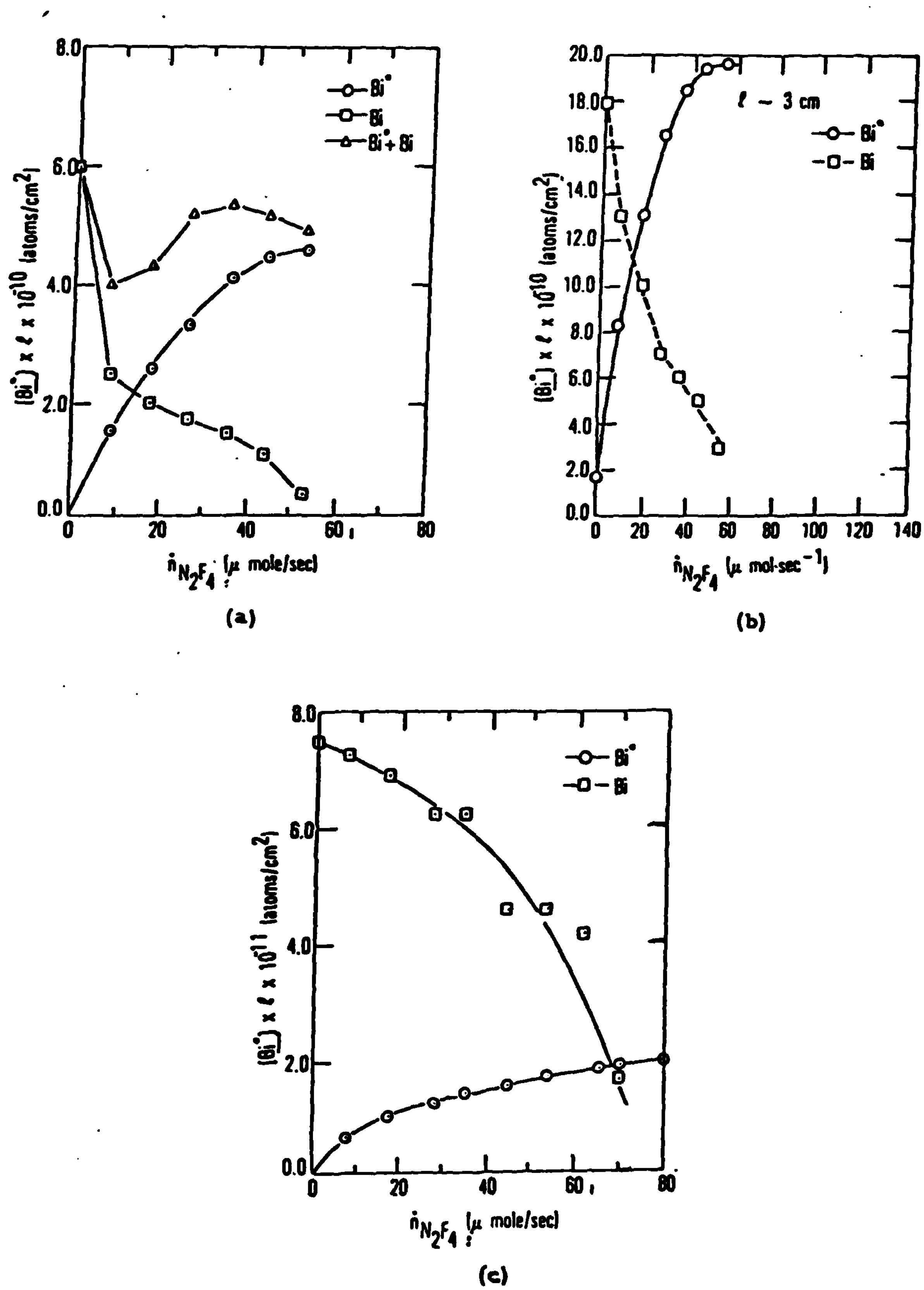

Fig. 5. Bi(6^4S) and Bi(6^2D) Concentrations Versus N_2F_4 Flow Rate at the Upstream Port. (a) Test 27A, (b) test 25, and (c) test 27B.

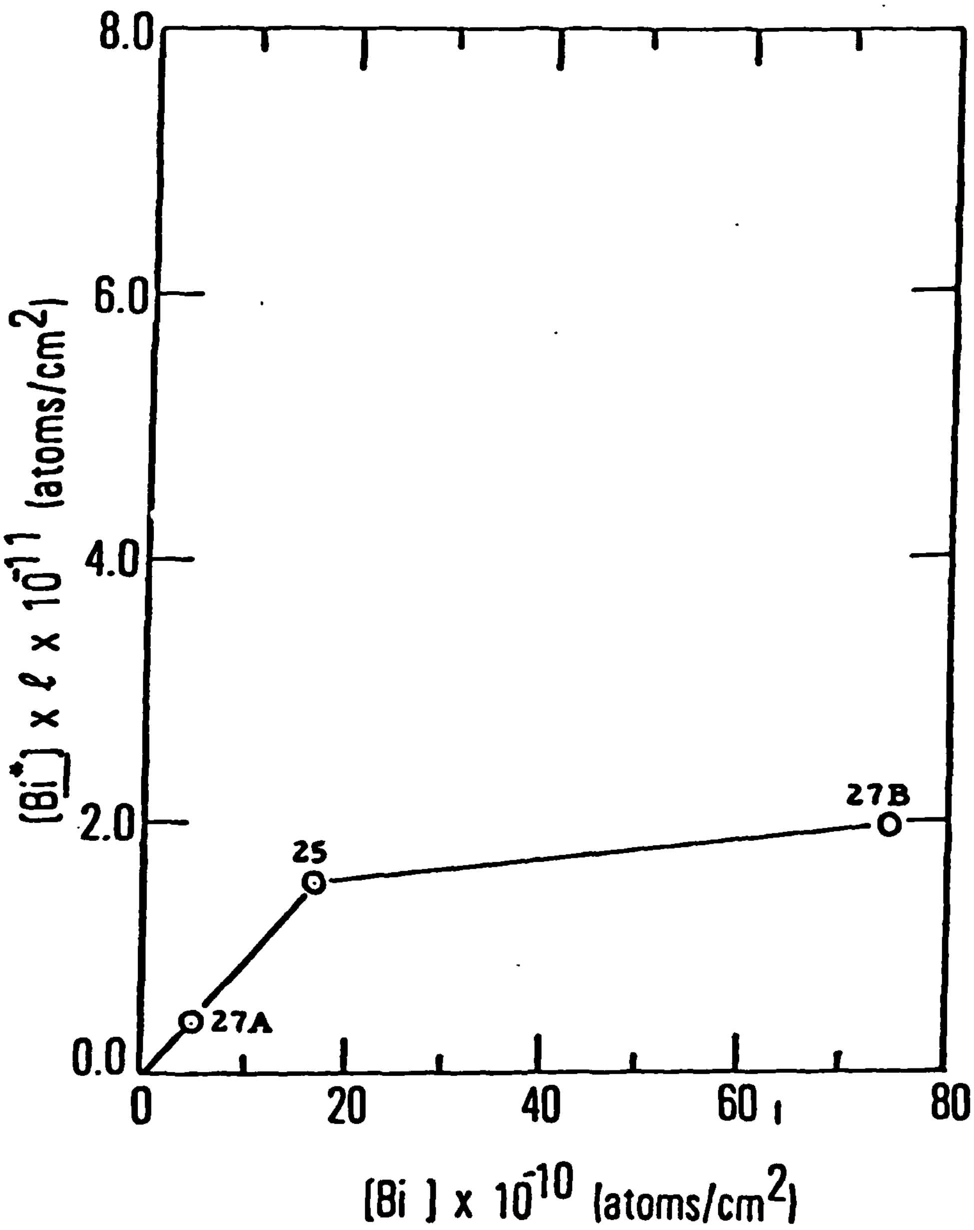

Fig. 6. $Bi(6^2D)$ Concentration Versus $Bi(6^4S)$ Concentration

four-fold increase in $Bi(6^4S)$ atoms only yields an additional 20% increase. This behavior is consistent with secondary reactions of $Bi(6^2D)$ with $NF(a^1\Delta)$.

This loss of $Bi(6^2D)$ is even more dramatically illustrated through observations at the downstream, 140 cm, port (Fig. 7). The large initial drop in the $Bi(6^4S)$ concentration results in a very small increase in $Bi(6^2D)$ concentration. Further, the rise in $Bi(6^2D)$ concentration is directly related to the rise in $Bi(6^4S)$ concentration.

All of the preceding observations are consistent with

$$(Bi)_2 + NF(a^1\Delta) \rightarrow (Bi)^{\ddagger}_2 \rightarrow 2Bi(6^4S) + NF(X^3\Sigma^-) \qquad (6)$$

as the secondary source of $Bi(6^4S)$ and

$$Bi(6^2D) + NF(a^1\Delta) \rightarrow BiF + N \qquad (7)$$

as a dominant removal process of the $Bi(6^2D)$.

Throughout our investigations, a blue glow was observed to fill the flow tube along the ~ 2-m length in a classic diffusion pattern from the metal heater. Scans in the 4000 to 5000 Å region produced the BiF(A-X) emission spectrum shown in Fig. 8. (The band heads are listed in Table 1.) This confirmed the presence of BiF in the flow. Studies are now in progress to further elucidate the reaction mechanisms involved in producing this blue emission.

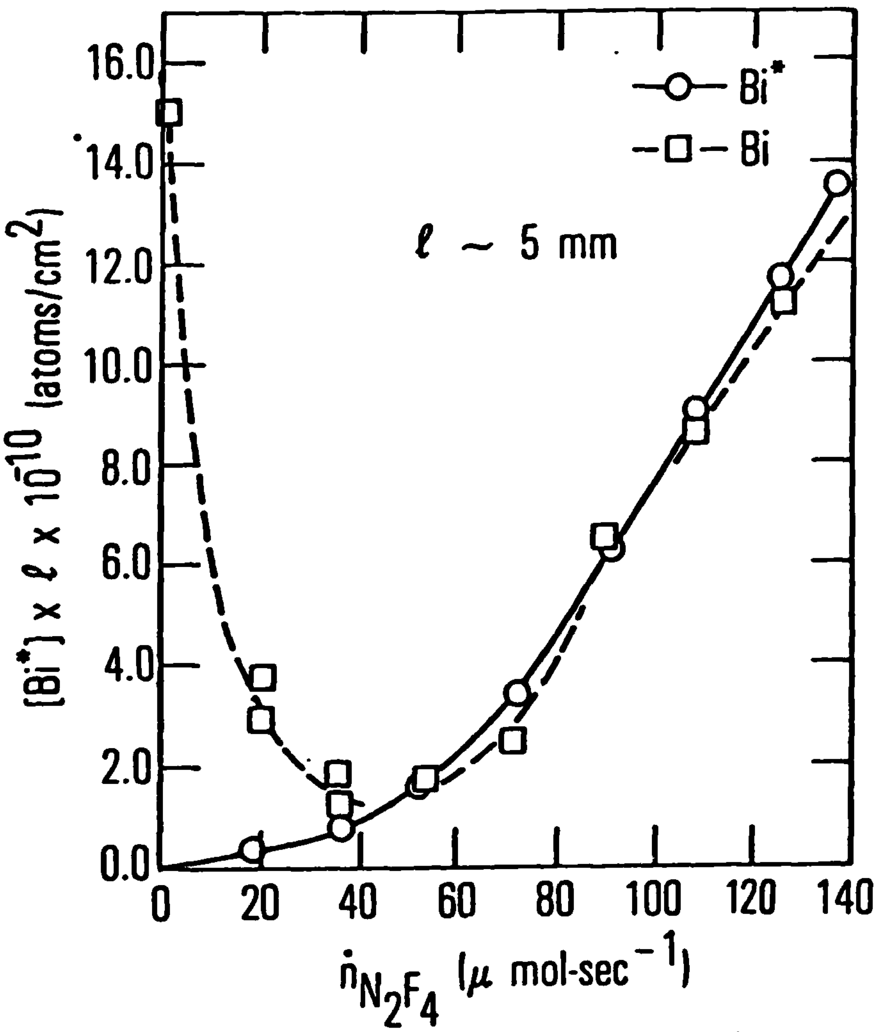

Fig. 7. Bi(6^4S) and Bi(6^2D) Concentrations Versus N_2F_4 Flow Rate at the Downstream Port

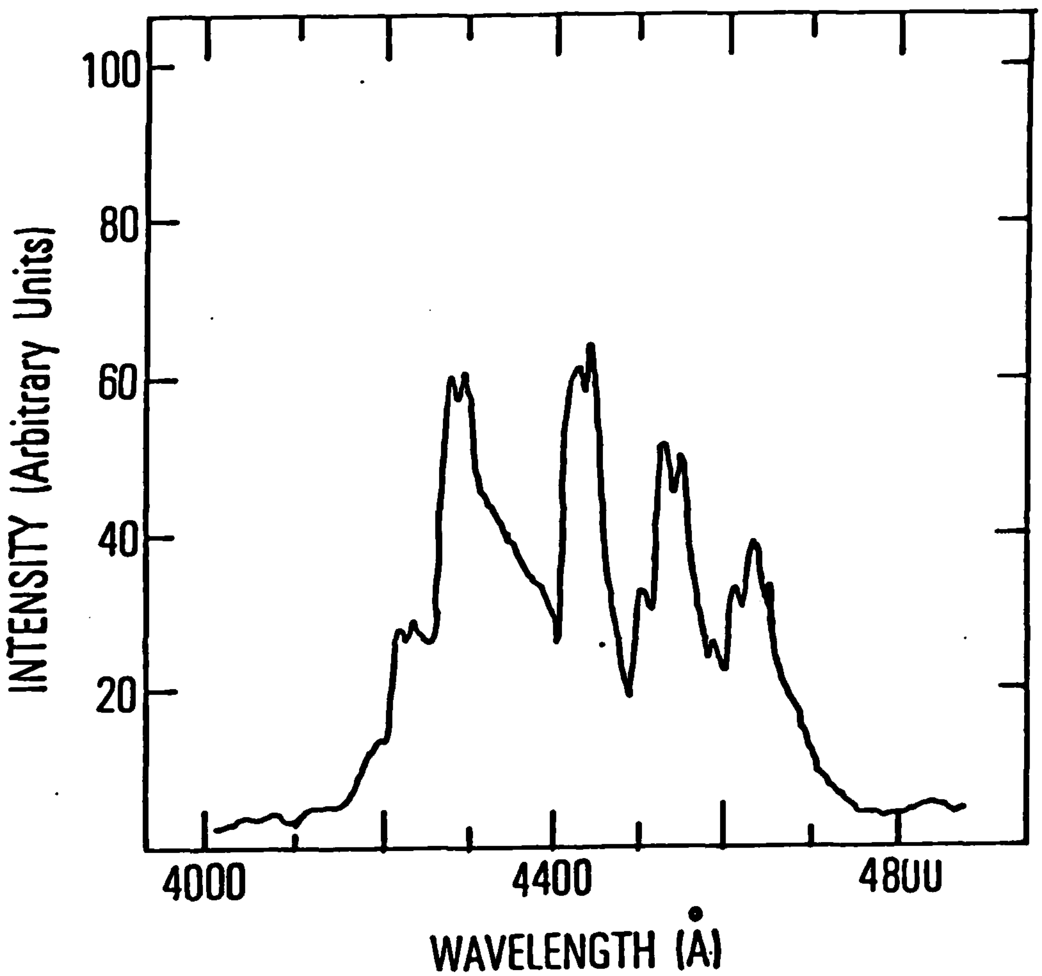

Fig. 8. Emission Spectrum of BiF(A-X)

Table 1. Principle Emission Spectra Within the 3000 to 9000 Å Region

Species	Transition	λ(Å)
$Bi(6^2D)$	$6^2D - 6^4S$	8758
$NF(a^1\Delta)$	$a^1\Delta - X^3\Sigma^-$	8742
$BiF(A^3\Sigma)$	$(A^3\Sigma, \nu') - (X^3\Sigma, \nu'')$	
	$\nu' - \nu''$	
	1 - 0	4232
	0 - 0	4368
	0 - 1	4467
	0 - 2	4570
	0 - 3	4677

IV. CONCLUSIONS

Studies have been conducted in a 10-cm flow-tube facility capable of producing $NF(a^1\Delta)$ in concentrations of about 10^{13} mol/cm^3. Into this medium was injected bismuth atoms at densities to 2.5×10^{11} atoms/cm^3, producing $Bi(6^2D)$ concentrations of about 7×10^{10} atoms/cm^3 at 8758 Å. Accompanying this was the corresponding reduction in the $Bi(6^4S)$ ground state resulting in inversions of greater than 90%.

While inversion has been shown, there appears to be a maximum level of $Bi(6^2D)$ achievable for the available $NF(a^1\Delta)$ concentrations caused by the existence of a loss of bismuth atoms. This loss mechanism is presently under investigation so that the potential of using this chemical system to power a high energy short wavelength laser system can be more reliably evaluated.

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